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## **Influence of Domain Structure on Magnetoresistance in Perovskite Manganite Grain Boundary Junctions**

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### **ABSTRACT**

We have been able to deduce a temperature dependence of the built-in potential in  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  grain boundary junctions. This has been performed by trimming a single grain boundary down to 1  $\mu\text{m}$  width with a focused ion-beam. We can thereby see the impact of single domain walls on the magnetoresistance and the current-voltage characteristics. We have also demonstrated the effect of averaging as we increased the number of junctions.

### **INTRODUCTION**

Since the discovery of colossal magnetoresistance (CMR) [1] in perovskite manganites these materials have attracted a lot of scientific attention. The interest is mainly due to their potential for technical applications, but also because of the many intriguing and complex properties. Grain boundaries (GBs) in manganites form magnetic junctions giving a tunnelling-like magnetoresistance response, both in granular films [2] and in bi-crystal films [3-7]. Bi-crystals have previously successfully been applied to study the colossal magnetoresistance of single manganite grain boundaries [3-7]. It has been shown that the Julliere model is not directly applicable to charge transport across manganite GBs [5,6] since the main origin of charge transport perpendicular to the grain boundary is not a direct tunnelling [4]. It has also been shown by magnetic force microscope that a grain boundary pins a magnetic domain wall [8].

The previous studies have focused on large-angle grain boundaries ( $\geq 24^\circ$ ), and the impact of single magnetic domains has previously not been thoroughly studied. Our previous study of noise in a multidomain structure [7] revealed an additional contribution originating from the GB region. Hence the motivation for our present work is to study the influence of junction width on its transport properties. This is achieved by decreasing the size of the grain boundary junctions (GBJs) in consecutive steps. Therefore we have studied the magnetoresistance (MR) and current-voltage (IV) dependence of a single low-angle GB as well as a GB array, and the dependences as function of width of the GBJ. This allows us to describe the magnetoresistance of single domain magnetic tunnel junctions.

### **EXPERIMENTAL DETAILS**

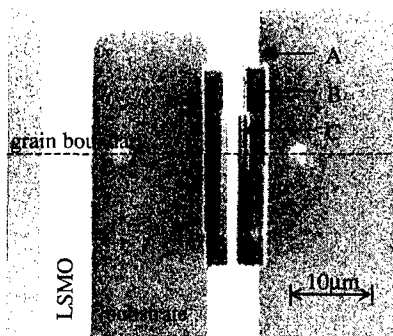
On a  $\text{LaAlO}_3$  bi-crystal substrate with symmetric misorientation angle of  $8.8^\circ$  a  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  film was grown by pulsed laser deposition. In this process a stoichiometric target was ablated by an excimer laser ( $\text{KrF}$ ,  $\lambda=248\text{nm}$ ) with an energy density of  $\sim 1.4\text{J}/\text{cm}^2$ . During deposition the substrate was held at  $740^\circ\text{C}$  in an oxygen pressure of 0.4mbar. A high degree of epitaxy of the 90nm-thick film was verified by x-ray  $\theta$ -2 $\theta$  and  $\phi$ -scans in a Bragg-Brentano diffractometer. The film was then patterned with photolithography and Ar-ion milling into a meander with 101 GB crossings, each 6 $\mu\text{m}$ -wide. Such a layout allows us to measure across one, two and three as well as one hundred GBJs. The MR properties were measured in a helium-cooled cryostat with a variable temperature insert and a

superconducting magnet. In all measurements the magnetic field was in the plane of the film and the MR was measured with a bias current of 10  $\mu$ A. The high-field MR has been deduced from the zero-field and field cooled resistance measurements, while the low-field MR was measured at specific temperatures.

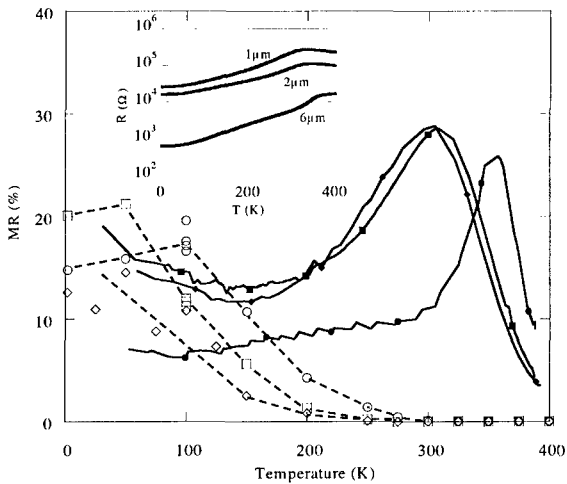
To study the behaviour of junction width we have trimmed one of the junctions. The trimming was performed in a focused ion-beam (FIB) with Ga-ion source. The junction was trimmed in two steps, the first FIB-process left a grain boundary junction 2  $\mu$ m wide and 20  $\mu$ m long, while in the second FIB-process the junction was trimmed down to 1  $\mu$ m width. The junction geometry is shown in figure 1.

## RESULTS

We estimate the temperature of the paramagnetic-ferromagnetic transition, the  $T_C$ , to be around 360K for a single 6  $\mu$ m wide grain boundary junction (figure 2). After trimming the junction the high-field MR as well as the resistance curves are shifted down with about 50K, which indicates a  $T_C$  just above 300K. This can be related to a heating or Ga contamination during the FIB-process. From figure 2 it is also clear that the onset of the grain boundary MR decreases as the GBJ is narrowed. As the junction is trimmed from 6  $\mu$ m to 2  $\mu$ m the onset decreases from 300K to 250K. Further trimming to 1  $\mu$ m results in a slight further decrease of the onset temperature. The temperature dependence of the grain boundary MR seems to be similar regardless of the width of the junction. The MR curves become smoother, as shown in figure 3a, when the number of GBs is increasing. The MR for a single GB is step-like, involving a few (5 to 10) resistance levels. Even though the effect is weak with the field perpendicular to the GB the step-like behaviour is clearly seen as the field is applied parallel to the GB. The effect of averaging, i.e. smoothing the MR by measuring several GBs, is therefore also most clear with the magnetic field parallel to the grain boundary. We note that the field for the MR peak does not change as more GBJs are included. On the other hand the maximum grain boundary magnetoresistance ( $MR^*$ ) decreases when increasing the number of GB included in the measurements. Trimming a grain boundary junction resulted in a decrease in the number of resistance-steps as illustrated in figure 3b. Roughly the average number of steps decreased from about 7 for the 6  $\mu$ m wide junction to 2-3 steps for the 2  $\mu$ m and 1  $\mu$ m wide junctions. The experiments did not reveal any specific temperature dependence for the number of resistance-steps, it is basically constant in the entire temperature range.



**Figure 1.** A secondary electron image of the trimmed junction after the second FIB-process. The grain boundary is indicated by the dashed line and the electrode is marked 'LSMO'. The width at 'A' is the original one-6  $\mu$ m, the width at 'B'-2  $\mu$ m and 'C' marks the trimmed '1  $\mu$ m' junction.



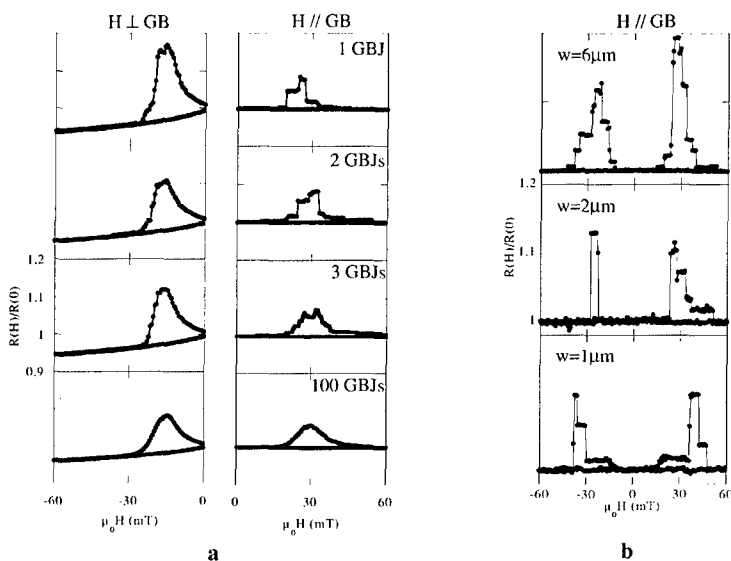
**Figure 2.** Temperature dependence of high-field (5T, solid lines) and low field ( $\sim H_c$ , open markers) magnetoresistance for a single GBJ with  $6\mu\text{m}$  (circles),  $2\mu\text{m}$  (boxes) and  $1\mu\text{m}$  (diamonds) width. At 100K the magnetoresistance was measured several times, this is indicated by several open markers. The dashed lines are guides for the eye. The inset shows the temperature dependence of the resistance for the GBJs.

## DISCUSSION

The grain boundary magnetoresistance effect in perovskite manganites is different from the colossal magnetoresistance observed in the same materials. In the kind of devices studied here both effects are present but have different temperature dependences. The grain boundary magnetoresistance has an onset at temperatures lower than the bulk Curie temperature and increases as the temperature falls. The IV-characteristics follows the same temperature dependence.

The resistance-area product of a manganite domain wall has previously been estimated to be about  $10^{-13}\Omega\text{m}^2$  [9]. For the  $1\mu\text{m}$  trimmed junction the cross section will be  $\sim 10^{-13}\text{m}^2$ , which results in a domain wall resistance of  $1\Omega$ . The latter is much smaller than the resistance measured in our experiments and hence the main voltage drop is due to dislocations or distortions at the GB. We believe, based on the results from the MR measurements (figure 3b), that we have been able to distinguish the influence of single domains on the MR and hence the impact of single domains on the IV characteristics of the junction.

In the IV-characteristics there seem to be two different regions. At low voltages the current goes as  $V^\alpha$  where  $\alpha=1-2$ , which is consistent with suggested quantitative models [4,6] for the current transport across the GB region: Gross et al [6] suggested a Glazman-Matveev [10] based theory which includes tunnelling through the GB region via localised states resulting in a voltage dependence of  $\alpha=1$  (plus extra terms of the order of  $\alpha=7/3$ ). On the other hand Todd et al [5] suggested to employ the Simmons model [11] which results in  $\alpha=1$  (plus extra terms of the order of  $\alpha=3$ ). In this voltage range we have too few data points to distinguish between



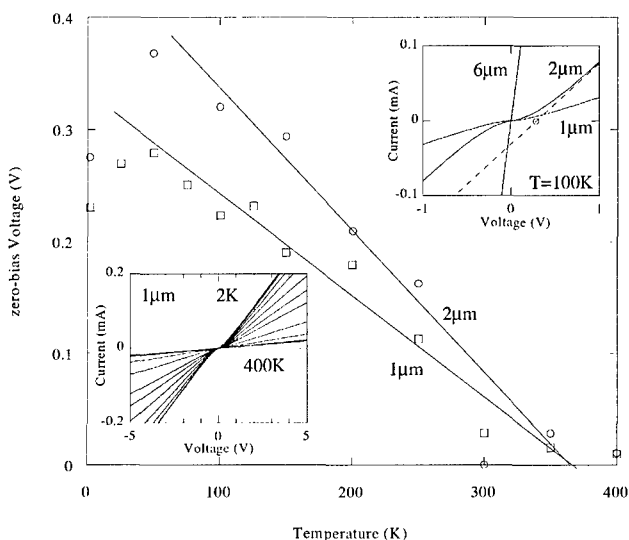
**Figure 3. a)** Magnetoresistance for 1, 2, 3 and 100 GBJ ( $6\mu\text{m}$  wide), the left column was measured with a magnetic field perpendicular to the grain boundary ( $H \perp \text{GB}$ ) while the right column displays the MR with a field parallel to the grain boundary ( $H // \text{GB}$ ). Since magnetoresistance is symmetric for positive and negative magnetic fields, only one part of the curve is displayed here. All plots are in the same scale. **b)** Single grain boundary magnetoresistance for the trimmed junction with 6, 2 and  $1\mu\text{m}$  width. Data for both subfigures are taken at 100K.

the suggested models. None of them is valid at high bias voltages where the voltage becomes comparable to the potential barrier.

For high voltages the IV-curve is linear up to 5V where the Joule heating effect starts to influence transport properties. This also suggests that the linear part can be extrapolated to zero bias-current that would give an estimate of the built-in potential,  $V_{bi}$ , discussed by Gross et al [6]. This assumption is valid at least within the error of a pre-factor. The measured potential and hence  $V_{bi}$  decreases down to zero at about the Curie temperature, see figure 4. This strengthens the argument that the non-linearity has a magnetic origin.

## CONCLUSIONS

By employing several as well as single grain boundary junctions we have been able to demonstrate the effect of magnetoresistance averaging in a granular manganite film. Then by trimming a single grain boundary junction we have been able to deduce the temperature dependence of the magnetic potential barrier. The latter differs from the temperature dependence of the grain boundary MR observed in the same sample. Thus, despite the fact that they both have magnetic origin, the non-linearity of IV-curves for manganite GBJs and the grain boundary MR does not describe the same physical effect.



**Figure 4.** Zero-bias voltage drops linearly with temperature to become zero at roughly the Curie temperature. The  $2\mu\text{m}$  and  $1\mu\text{m}$  wide junctions are represented by open circles and open boxes respectively, the lines are guides for the eye. The upper inset show the IV-characteristics at 100K for the  $6\mu\text{m}$ ,  $2\mu\text{m}$  and  $1\mu\text{m}$  wide junctions. The zero-bias voltage is deduced from the linear part (indicated by a dashed line and a circle at zero current). The influence of temperature on the IV-characteristics is shown in the lower inset with IV-curves at 2K, 25K, 50K, 75K, 100K, 125K, 150K, 200K, 250K, 300K, 350K, 400K.

## ACKNOWLEDGEMENTS

The work has been supported by The Swedish Research Council (TFR) and The Board for Strategic Research (SSF) with programs "OXIDE" and "Transport in mesoscopic structures".

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